

Sussex Research Online

Laminar flame characteristics of natural gas and dissociated methanol mixtures diluted by nitrogen

Article (Accepted Version)

Zhu, Zhennan, Liang, Kun, Zeng, Dongjian, Meng, Zhongwei, Zuo, Zinong and Pei, Yiqiang (2019) Laminar flame characteristics of natural gas and dissociated methanol mixtures diluted by nitrogen. Journal of the Energy Institute. ISSN 1743-9671

This version is available from Sussex Research Online: <http://sro.sussex.ac.uk/id/eprint/82439/>

This document is made available in accordance with publisher policies and may differ from the published version or from the version of record. If you wish to cite this item you are advised to consult the publisher's version. Please see the URL above for details on accessing the published version.

Copyright and reuse:

Sussex Research Online is a digital repository of the research output of the University.

Copyright and all moral rights to the version of the paper presented here belong to the individual author(s) and/or other copyright owners. To the extent reasonable and practicable, the material made available in SRO has been checked for eligibility before being made available.

Copies of full text items generally can be reproduced, displayed or performed and given to third parties in any format or medium for personal research or study, educational, or not-for-profit purposes without prior permission or charge, provided that the authors, title and full bibliographic details are credited, a hyperlink and/or URL is given for the original metadata page and the content is not changed in any way.

Accepted Manuscript

Laminar Flame Characteristics of Natural Gas and Dissociated Methanol Mixtures Diluted by Nitrogen

Zhennan Zhu, Kun Liang, Dongjian Zeng, Zhongwei Meng, Zinong Zuo, Yiqiang Pei



PII: S1743-9671(19)30038-8

DOI: <https://doi.org/10.1016/j.joei.2019.03.002>

Reference: JOEI 573

To appear in: *Journal of the Energy Institute*

Received Date: 8 January 2019

Revised Date: 1 March 2019

Accepted Date: 4 March 2019

Please cite this article as: Z. Zhu, K. Liang, D. Zeng, Z. Meng, Z. Zuo, Y. Pei, Laminar Flame Characteristics of Natural Gas and Dissociated Methanol Mixtures Diluted by Nitrogen, *Journal of the Energy Institute*, <https://doi.org/10.1016/j.joei.2019.03.002>.

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Laminar Flame Characteristics of Natural Gas and Dissociated Methanol Mixtures Diluted by Nitrogen

Zhennan Zhu ^a, Kun Liang ^{b*}, Dongjian Zeng ^{a*}, Zhongwei Meng ^a, Zinong Zuo ^a, Yiqiang Pei ^c

^a School of Automobile and Transportation, Xihua University, Chengdu 610039, PR China

^b Department of Engineering and Design, University of Sussex, Brighton BN1 9QT, UK

^c State Key Laboratory of Engines, Tianjin University, Tianjin 300072, PR China

* Corresponding author. Tel: +44 1273 678573. Email address: kun.liang@sussex.ac.uk

Tel: +86 135 5862 1686. Email address: zdj.640102@163.com

Abstract: The effect of dissociated methanol ($H_2:CO=2:1$ by volume) on laminar burning velocity of natural gas (methane as the main component) was studied by using a constant volume bomb (CVB). Nitrogen, as diluent gas, was added into the natural gas (CH_4) - dissociated methanol (DM) mixtures to investigate the dilution effect. Experiments were conducted at initial temperature of 343 K and initial pressure of 0.3 MPa with equivalence ratios from 0.8 to 1.4. Laminar burning velocities were calculated through Schlieren photographs, correlation of in-cylinder pressure data and Chemkin-Pro. Results show an increase in laminar burning velocity with initial temperature and proportion of dissociated methanol but a decrease with initial pressure and proportion of nitrogen. The laminar burning velocities were 25.1 cm/s, 38.7 cm/s and 83.2 cm/s respectively at stoichiometric ratio when the proportions of the dissociated methanol were 0%, 40% and 80%. Adding more dissociated methanol tends to shift the peak burning velocity towards the richer side while adding nitrogen has the opposite effect. More dissociated methanol will lead to earlier cellularity.

Keywords: Natural gas, Dissociated methanol, Nitrogen, Laminar burning velocity, Cellularity

Nomenclatures

DM	dissociated methanol	ρ_b	density of burned gas, kg/m^3
LM	liquid methanol	Le_{eff}	effective Lewis number
N	nitrogen	λ	thermal conductivity, $W/(m \cdot K)$
EGR	exhaust gas recirculation	C_p	specific heat capacity, $J/(kg \cdot K)$
DAQ	data acquisition	D_T	thermal diffusion coefficient, m^2/s
CVB	constant volume bomb	D_m	mass diffusion coefficient, m^2/s
R	instantaneous flame radius, cm	DM0	0% dissociated methanol by volume
S_n	stretched flame propagation speed, cm/s	DM40	40% dissociated methanol by volume
S_l	unstretched flame propagation speed, cm/s	DM80	80% dissociated methanol by volume
S_u	laminar burning velocity, cm/s	DM40N5	40% dissociated methanol and 5% nitrogen by volume
K	stretch rate, $1/s$	DM40N10	40% dissociated methanol and 10% nitrogen by volume
L_b	Markstein length, cm	DM40N15	40% dissociated methanol and 15% nitrogen by volume
ρ_u	density of unburned gas, kg/m^3		

1. Introduction

The fossil fuel resource shortages and environmental pollutions lead to requirement of alternative fuels for utilizing in spark-ignition (SI) engines, particularly in China, where the automobile market is now the largest in the world. Natural gas, which mainly consists of methane (CH_4), is regarded as a type of alternative fuels because of its sufficiency in the world (149.76 trillion cubic meters) [1] and eco-friendly combustion properties. However, the flame propagation speed is a shortcoming. At ambient temperature and pressure, the flame propagation speed of natural gas-air mixture is about 40 cm/s at stoichiometric ratio. In order to reach a higher flame propagation speed with lower ignition energy, hydrogen (H_2) can be added into natural gas [2]. The addition of H_2 can also reduce the toxic emissions such as hydrocarbon (HC), improve the thermal efficiency and extend the combustible limit [3, 4 and 5]. However, the main concerns about hydrogen are the high cost and very strong chemical reactivity.

Syngas, consisting primarily of hydrogen (H_2) and carbon monoxide (CO), can be used to replace hydrogen due to the superiority of being produced easily [6]. Methanol, due to its mature production technology and rich source of raw materials, is regarded as promising alternative fuel, which can be used to produce syngas. Methanol is usually made from naphtha, yellow phosphorus tail gas, and natural gas [7]. A simple way to produce dissociated methanol for use in combustion engines is to dissociate methanol under the effect of heat and catalyst according to Eq.(1) as the reaction temperatures are relatively low [8]. Fig.1 shows the diagram of the dissociated methanol engine.

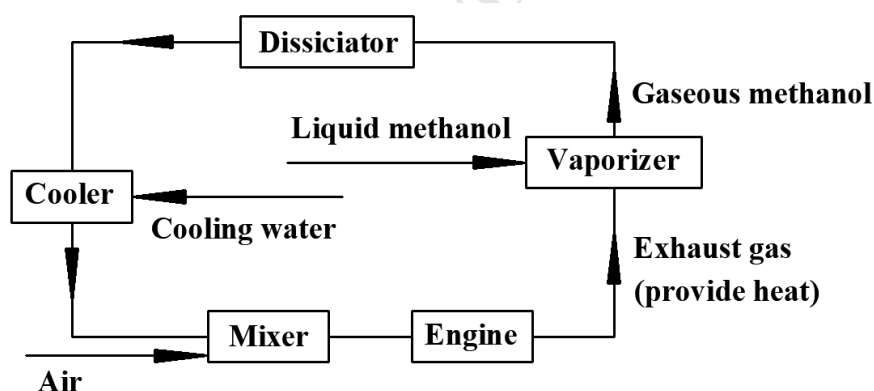


Fig. 1 Diagram of the dissociated methanol engine [7]

The low heating value of DM is 20% higher than liquid methanol (LM), so that the chemical energy can be further increased after being dissociated, which will result in higher thermal efficiency. China, with vast coal reserves, produces large amount of methanol every year, over 90% of which comes from coal [9]. It is feasible to use dissociated methanol as a type of alternative fuels in combustion engines. However, in-cylinder temperature also increases when adding dissociated methanol into natural gas [10], which will increase the emission of NO_x . Adding diluent gases, which can function as exhaust gas recirculation (EGR), is a feasible way to solve this problem since combustion temperature can be decreased by increasing the specific heat capacity of the mixtures and hindering the chemical reaction [11]. Nitrogen (N_2), an important component of EGR gas, is of great research value and has been investigated in many studies [5, 12 and 13].

To date, studies on natural gas, syngas and diluent gas are common, but most of them focus on the effects of dilution on each of the other two [12 and 13]. Studies on the natural gas-syngas engine mainly focus on the engine performance, but these works are relatively lack of details in burning characteristics [14 and 15]. Studies on the effect of dilution on the laminar flame of natural gas-syngas mixture can hardly be found.

Laminar burning velocity, which is closely related to burning duration, power output and efficiency [16 and 17], is one of the most important parameters for all types of fuels. Laminar burning velocity can also be used to predict turbulent burning velocity, to validate combustion kinetics model and to help with design of combustion system. However, modelling of burning velocity is very difficult as it requires valid mechanisms [18]. There are many methods of measuring laminar burning velocity, of which constant volume bomb method is the most versatile [38].

The present study focuses on the laminar burning velocities of natural gas-dissociated methanol mixtures and the effect of adding nitrogen. A constant volume bomb was adopted along with Schlieren photography technique. The laminar burning velocities have been calculated from both in-cylinder pressure data and Schlieren photographs. Numerical simulations using Chemkin-Pro with the latest San Diego chemical-kinetic mechanism were undertaken to predict the laminar burning velocity.

1. Experimental Setup and Data Processing

1.1. Experimental Setup

A constant volume bomb (CVB, shown in Fig. 2) was used to conduct experiments of premixed combustion of natural gas-DM-air mixtures and natural gas-DM-nitrogen-air mixtures. The constant volume bomb was heated by electrical heating tape entwined around and was wrapped by heat-insulation wool to reduce the heat loss. Initial temperature can be maintained by a proportional-integral-derivative (PID) controller and K-type thermocouple. The Schlieren photography system contained a tungsten-halogen lamp (100 W), two concave mirrors (focal length 110 mm), two plane mirrors and a high-speed camera (Phantom V7.3, photo frequency of 10,000 fps, image resolution of 512×512 pixels). A triggering control unit (ECTEK Calibration V2) was used to start the ignition, photographing and data acquisition at the same time. A pressure sensor (KISTLER 6125C) was adopted along with data acquisition card (DAQ, NI USB-6365, sampling frequency 100 KHz) and a charge amplifier (KISTLER 5018A). Both pressure data and photographs were recorded during combustion. Table 1 shows the parameters of the constant volume bomb. The volume of the bomb is 22.4 L and the optical window has a diameter of 120 mm.

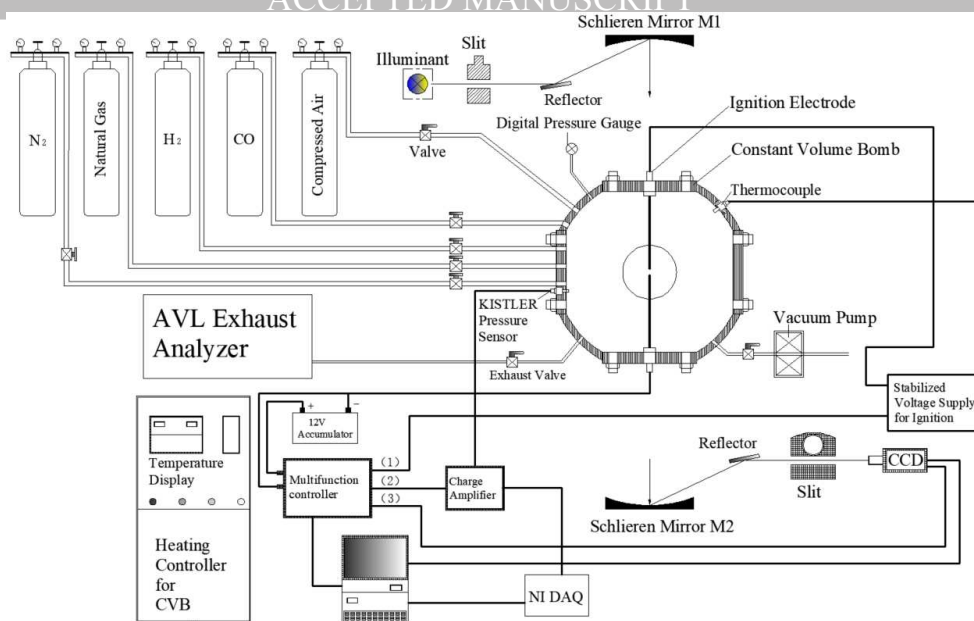


Fig. 2 Experimental setup consisting of a constant volume bomb, a Schlieren system, an intake system, a vacuum pump, an exhaust analyzer, a temperature controlling system and a data acquisition system.

Table 1 Parameters of the constant volume bomb

Parameters	Value
Inner diameter (mm)	350
Volume (L)	22.4
Maximum heating temperature (K)	600
Maximum pressure (MPa)	4
Effective diameter of windows (mm)	120
Ignition electrode gap (mm)	2
Ignition pulse width (ms)	2
Ignition primary voltage (V)	14

Table 2 shows the test conditions of laminar burning of premixed natural gas–DM–air mixtures with nitrogen as diluent gas. The natural gas–DM mixtures with DM volume fraction of 0%, 40% and 80% are named as DM0, DM40, and DM80 respectively. For DM40, the mixtures with nitrogen fraction of 5%, 10% and 15% are named as DM40N5, DM40N10 and DM40N15 respectively.

Table 2 Test conditions of premixed natural gas–DM–air mixtures with nitrogen as diluent gas

Parameters	Value
Initial temperature (K)	343
Initial pressure (MPa)	0.3
Equivalence ratio	0.8–1.4
DM fraction	0% (DM0), 40% (DM40), 80% (DM80)
N ₂ fraction	0%, 5% (N5), 10% (N10), 15% (N15)

Note that DM fraction means the volume fraction of DM in natural gas-DM mixtures, and N_2 fraction means the volume fraction of N_2 in natural gas-DM-air- N_2 mixtures, which are calculated as below:

$$DM \% = (V_{H_2} + V_{CO}) / (V_{H_2} + V_{CO} + V_{CH_4}) \quad (2)$$

$$N_2 \% = V_{N_2} / (V_{H_2} + V_{CO} + V_{CH_4} + V_{N_2} + V_{air}) \quad (3)$$

1.2. Data Processing

The schematic of the data processing for the laminar burning velocity measurement is shown in Fig.3. The processing of the Schlieren photography includes the extraction of flame radius (b) and detection of cellularity. The processing of the in-cylinder pressure data (LabVIEW data, shown as (a)) using *BOMB* program is shown in the blue charts and the result from correlation is shown in (d).

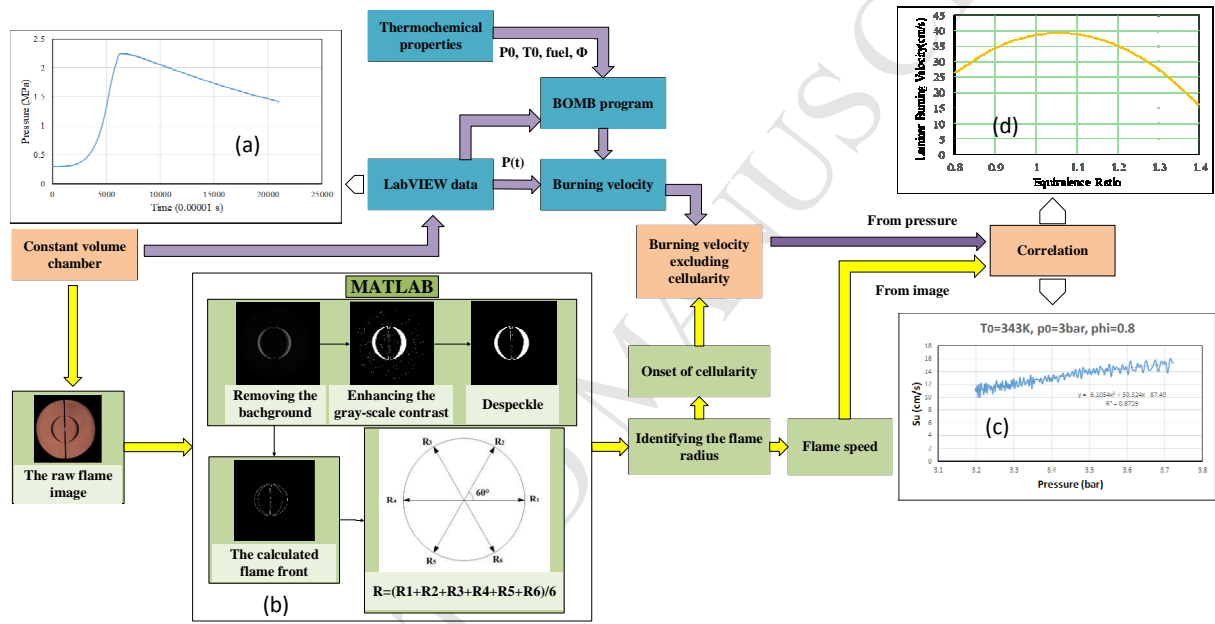


Fig. 3 Schematic of data processing from both in-cylinder pressure and Schlieren photographs.

1.2.1. Schlieren Photography

A MATLAB code was written to compute the outwardly propagating flame radius R from the flame images. The range of chosen radii is 6~25mm, since the effect of ignition energy can be neglected when $R > 6$ mm [19] and the pressure almost stabilized when $R < 25$ mm [20]. The raw flame images with $6 \text{ mm} < R < 25 \text{ mm}$ were collected in this work, and the range of flame radii was adjusted according to the onset of the cellularity.

The stretched flame propagation speed (S_n) can be expressed as below:

$$S_n = dR / dt \quad (4)$$

in which t is the time from ignition.

The flame stretch rate (K) is the instantaneous rate of the change in a flame surface element of area A , which is calculated according to:

$$K = d(\ln A) / dt = 2S_n / R \quad (5)$$

The extraction of the unstretched flame speed (S_l) and Markstein length (L_b) can be calculated based on a linear model:

$$S_l - S_n = L_b K \quad (6)$$

Chen [21] concluded that Eq. (6) is not accurate enough and two non-linear models for the extraction of S_l could be adopted:

$$S_n = S_l - 2S_l L_b / R \quad (7)$$

$$\ln(S_n) = \ln(S_l) - 2S_l L_b / R S_n \quad (8)$$

Eq.(7) was proposed by Markstein [22] and Eq. (8) was proposed by Kelley and Law [23].

Lewis number (L_e), is essential when choosing linear or non-linear models. Lewis number can be calculated according to

$$L_e = D_T / D_m = \lambda / \rho_u C_p D_m \quad (9)$$

in which λ is the heat conductivity; ρ_u is the density of unburned gas; C_p is the specific heat capacity; D_T is the thermal diffusion coefficient; D_m is the mass diffusion coefficient of insufficient reactant (fuel at lean side and oxygen at rich side) to inert gas (mostly nitrogen). The effective Lewis number of blended gas can be obtained by

$$Le_{eff} = \sum_{i=1}^n x_i Le_i \quad (10)$$

in which x_i is the i^{th} component's volume fraction; Le_i is the i^{th} component's Lewis number.

The non-linear model shown in Eq.(7) is accurate when $L_e > 1$ (or $L_b > 0$) while the non-linear model shown in Eq.(8) is accurate when $L_e < 1$ (or $L_b < 0$) according to [21]. The linear model is suitable when Lewis number is close or equal to 1 (the Markstein length is close or equal to 0) [24].

The laminar burning velocity (u_l) is calculated as below

$$S_u = S_l (\rho_b / \rho_u) \quad (11)$$

in which ρ_b is the density of burned gases.

An accurate ruler was used to estimate the deviation of spherical flame radius. The uncertainty of the flame radius was proved to within the range of ± 0.25 mm. The constant volume bomb used in this work was large enough so that the flame deformation can be avoided [25].

Flame radiation, which will also inherently influence the extraction of unstretched flame propagation speed, is another factor that should be taken into account. Yu et al. [26] proposed the following implicit expression to evaluate the effect of flame radiation

$$u_{l,RCFS} - u_{l,EXP} = 0.82 u_{l,EXP} (u_{l,EXP} / S_0)^{-1.44} (T_u / T_0) (P_u / P_0)^{-0.2} \quad (12)$$

in which $S_0 = 1$ cm/s, $T_0 = 298$ K, $P_0 = 0.1$ MPa, and $u_{l,RCFS}$ and $u_{l,EXP}$ are radiation-corrected laminar burning velocity and experimental laminar burning velocity respectively. In this study, the difference between $u_{l,RCFS}$ and $u_{l,EXP}$ was less than 0.5 cm/s (negligible).

Considering N pairs of the computed values of $(K, S_n)_i$, Markstein length should be calculated as below

$$L_b = -\sum_{i=1}^N (K_i - \bar{K})(S_{ni} - \bar{S}_n) / \sum_{i=1}^N (K_i - \bar{K})^2 \quad (13)$$

in which $\bar{S}_n = \frac{1}{N} \sum_{i=1}^N S_{ni}$ and $\bar{K} = \frac{1}{N} \sum_{i=1}^N K_i$. The standard error of one operating point can be estimated as the following equation according to [27]

$$e = \sqrt{\frac{1}{N-2} \left[\sum_{i=1}^N (S_{ni} - \bar{S}_n)^2 + L_b \sum_{i=1}^N (K_i - \bar{K})(S_{ni} - \bar{S}_n) \right]} \quad (14)$$

The uncertainty of laminar burning velocity based on equations above is calculated to be 0.8 cm/s, and the experimental uncertainty of the unstretched flame propagation speed in present work is less than 5%. The laminar burning velocity data calculated from Schlieren photographing is named as ‘Schlieren’ when they are compared with other two methods.

1.2.2. Pressure Trace

The *BOMB* program [28] is a multi-zone numerical model which was used to solve the conservation equations for mass and energy and the composition of ten major combustion products in each zone. In this model, the unburnt gas has a uniform temperature, pressure and composition. The pressure is assumed to be uniform across the bomb, and the gas is assumed to behave semi-perfectly, which means the gas can satisfy the ideal gas law but it is viscous.

The laminar burning velocity was calculated by combining the experimental pressure data recorded by LabVIEW and the zone file output from the *BOMB* program [29]:

$$S_u = \frac{dr_i}{dt} \left(\frac{r_i}{r_b} \right)^2 \left(\frac{p}{p_i} \right)^{\frac{1}{\gamma_u}} \quad (15)$$

where r_i is the initial radius of the current mass fraction burned prior to combustion, r_b is the flame radius, γ_u is the ratio of specific heats of the unburned mixture, and p_i is the initial pressure.

The initial radius r_i depends on the mass fraction burned as follow

$$r_i = \sqrt[3]{xR^3} \quad (16)$$

where x is the mass fraction of burned gas and R is the radius of the constant volume bomb.

Quadratic polynomials were adopted for correlating the laminar burning velocities from pressure data. The laminar burning velocity data from this method is named as ‘Correlated’ in comparison with two other methods in Section 4.

2. Numerical Model

The steady-state, one-dimensional freely propagating laminar flame model in Chemkin Pro was used to predict the laminar burning velocities of the premixed natural gas-DM-air mixtures with diluent. The adiabatic laminar flame speed was determined from the heat flux method so that no radiative heat loss was considered. The hybrid time-integration/Newton-iteration technique with adaptive meshes and mixture-averaged transport parameters was applied to solve the steady-state mass, species and energy conservation equations of the flames. The mechanism adopted is San Diego mechanism [30], which is a small detailed mechanism developed for the combustion of hydrocarbon fuels in air or oxygen-inert mixtures. The mechanism has 268 reactions and 54 species [31]. This mechanism has been validated [32]. Table 3 shows the

simulation conditions for the premixed mixtures. The burning velocity data from simulation is named as 'Chemkin' for comparison with other two methods.

Table 3 Simulation conditions of laminar flame for premixed natural gas-DM-air mixtures with nitrogen as diluent

Parameters	Value
Initial temperature (K)	300, 343, 350, 450
Initial pressure (MPa)	0.1, 0.3, 0.5
Equivalence ratio	0.8-1.4
DM fraction	0% (DM0), 40% (DM40), 80% (DM80)
N ₂ fraction	0%, 5% (N5), 10% (N10), 15% (N15)

3. Results and Discussions

4.1. Effective Lewis number

The effective Lewis numbers of natural gas-DM-air mixtures and diluted mixtures are shown in Fig.4 and Fig.5 respectively. Effective Lewis number is also an essential index for diffusion instability. High effective Lewis number will suppress preferential diffusion instability. Fig.4 and Fig.5 show an increase in effective Lewis number with equivalence ratio, indicating that the richer the mixture is, the more stable the flame is. Fig.4 shows a decrease in effective Lewis number with increasing fraction of dissociated methanol at lean side, but an increase at rich side. The reason is that the thermal diffusivity increases with equivalence ratio, and the mass diffusivity of natural gas-DM mixtures to nitrogen is higher than that of oxygen to nitrogen. Fig.5 shows a decrease in effective Lewis number with an increasing fraction of nitrogen due to a decrease in thermal diffusivity. Therefore, preferential diffusion instability of natural gas-DM-air flame will be enhanced when adding dissociated methanol at lean side but will be suppressed at rich side, and it will be enhanced when adding nitrogen at both lean and rich sides.

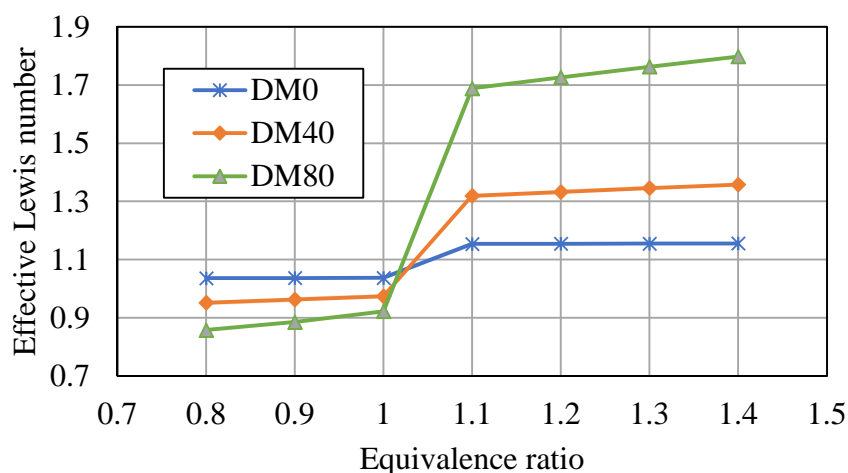


Fig. 4 Effective Lewis number of natural gas-DM mixtures at 343 K and 0.3 MPa

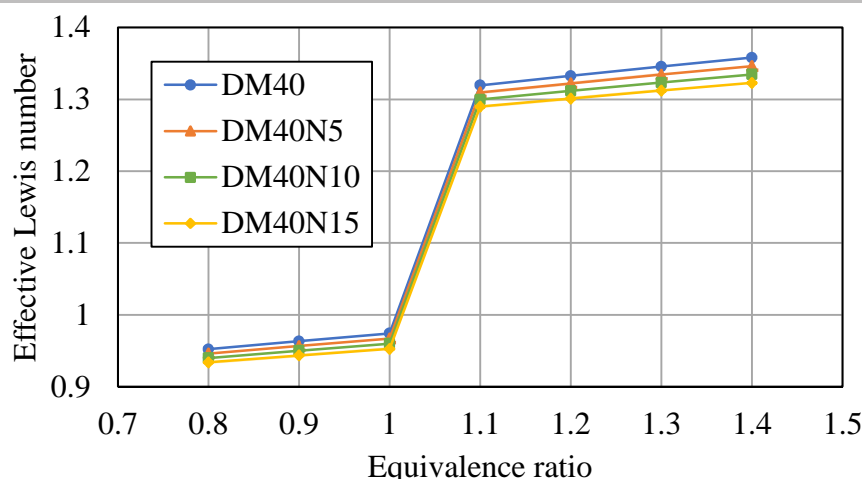


Fig. 5 Effective Lewis number of natural gas with 40% dissociated methanol (DM40) diluted by nitrogen at 343 K and 0.3 MPa

4.2. Natural gas

Fig.6 shows the laminar burning velocities of pure natural gas (DM0) from correlating pressure data, Schlieren and Chemkin. The correlated data in present work agrees with results from Nathan and Stone [33] who also adopted in-cylinder pressure data and constant volume bomb method under wide range of temperatures and pressures. The peak burning velocity occurs close to stoichiometric ratio. Both Chemkin and Schlieren photography give higher laminar burning velocities than correlating pressure data, with maximum difference of 5 cm/s.

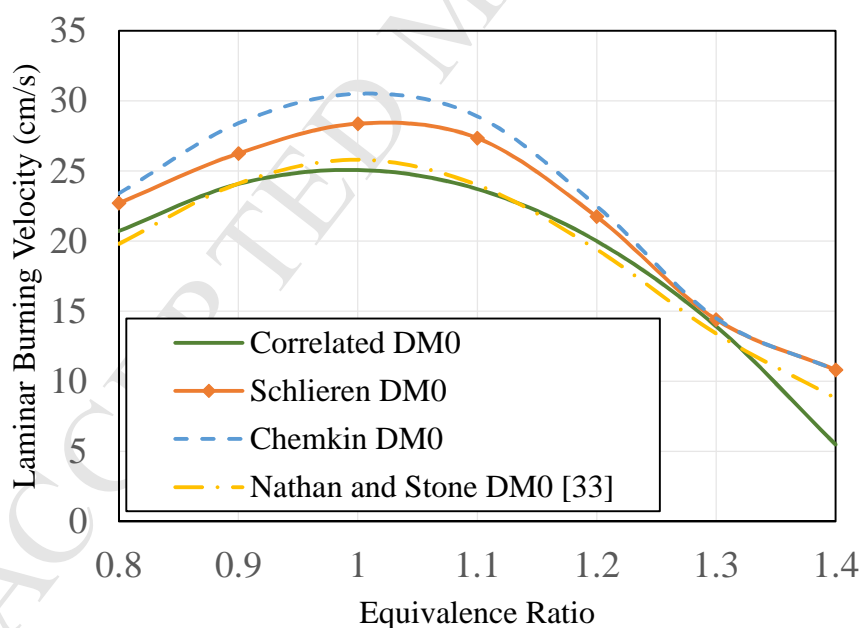


Fig. 6 Burning velocities of natural gas without dissociated methanol (DM0) from correlation, Schlieren, and Chemkin at 343 K and 0.3 MPa in comparison with literature data

Fig.7 shows the laminar burning velocities of pure natural gas from Chemkin at initial pressure of 0.1 MPa and different initial temperatures while Fig.8 shows the laminar burning velocities of pure natural gas from Chemkin at initial temperature of 450 K and different initial pressures. It is clear that burning velocity increases with temperature but decreases with pressure.

Peak burning velocity occurs at slightly richer equivalence ratio than stoichiometric mixture (equivalence ratio of 1.05), which agrees with previous studies [11, 19 and 33] at ambient pressure.

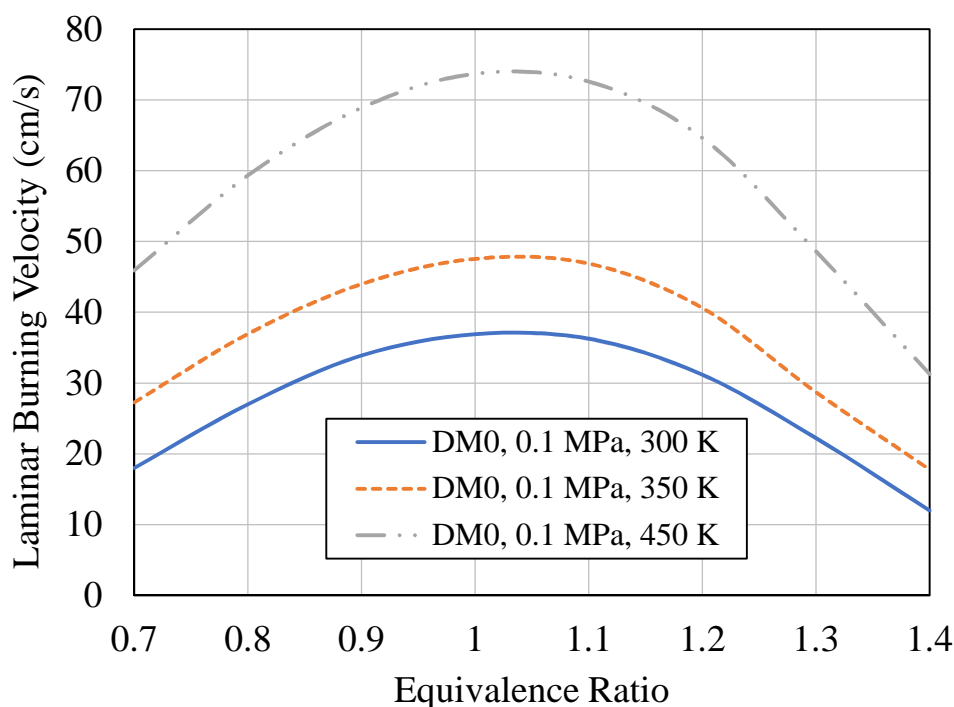


Fig. 7 Burning velocity of natural gas without dissociated methanol (DM0) from Chemkin at different temperatures and 0.1 MPa

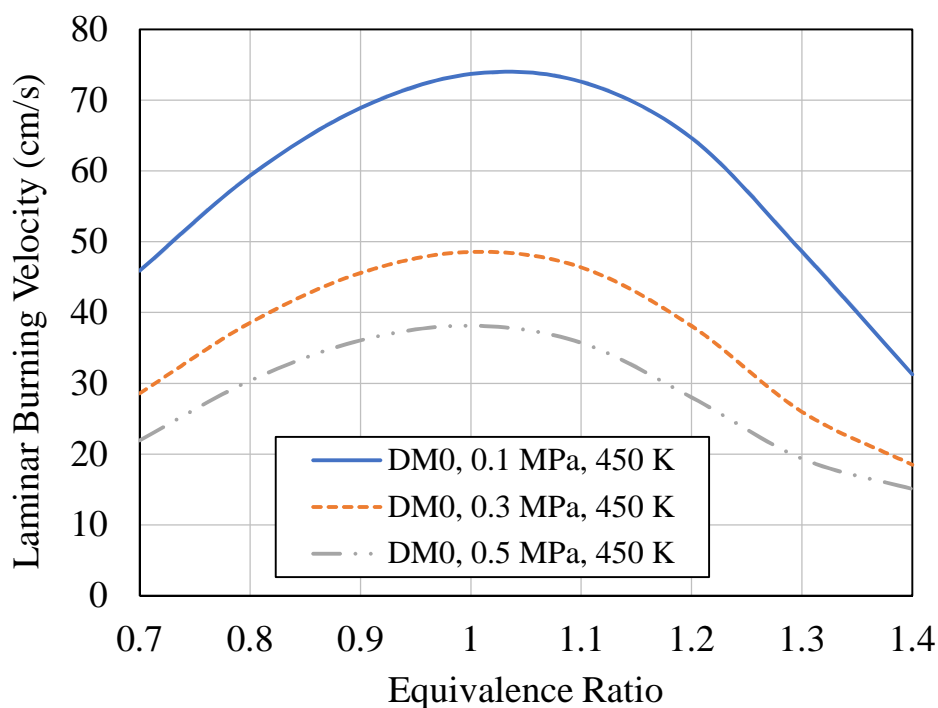


Fig. 8 Burning velocity of natural gas without dissociated methanol (DM0) from Chemkin at different pressures and 450 K

4.3. Natural gas-Dissociated Methanol Mixture

Laminar burning velocities of natural gas with 40% dissociated methanol added (DM40) and

80% dissociated methanol added (DM80) obtained from three methods are shown in Fig.9 and Fig.10 respectively. Fig.9 shows that laminar burning velocities from three methods are very close to each other. Laminar burning velocity from Chemkin is generally higher than that from correlation and Schlieren, but the difference among them is negligible (3 cm/s for DM40 and 5 cm/s for DM80 at most). Schlieren and correlation show agreement with each other.

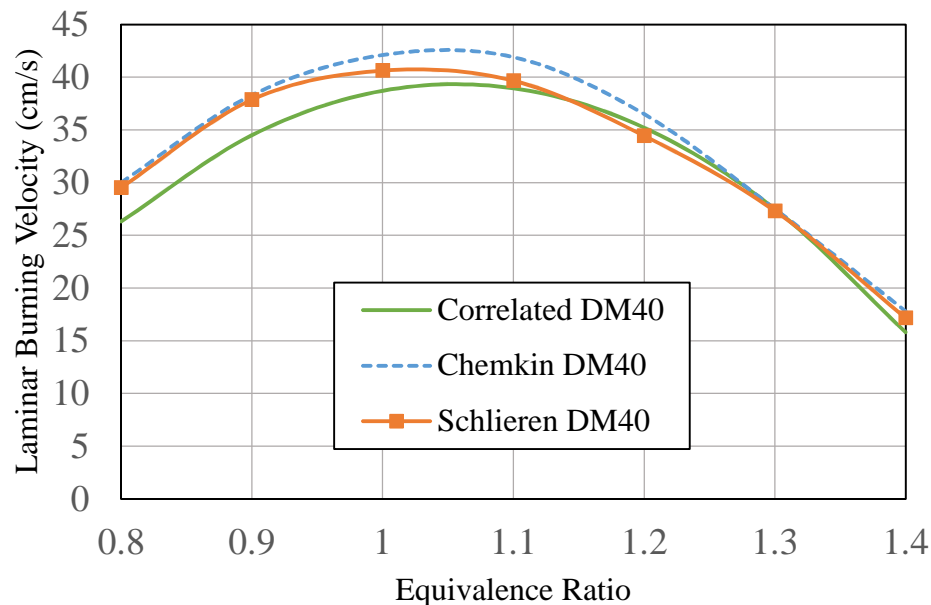


Fig. 9 Burning velocity of natural gas with 40% dissociated methanol (DM40) from correlation, Chemkin and Schlieren at 343 K and 0.3 MPa

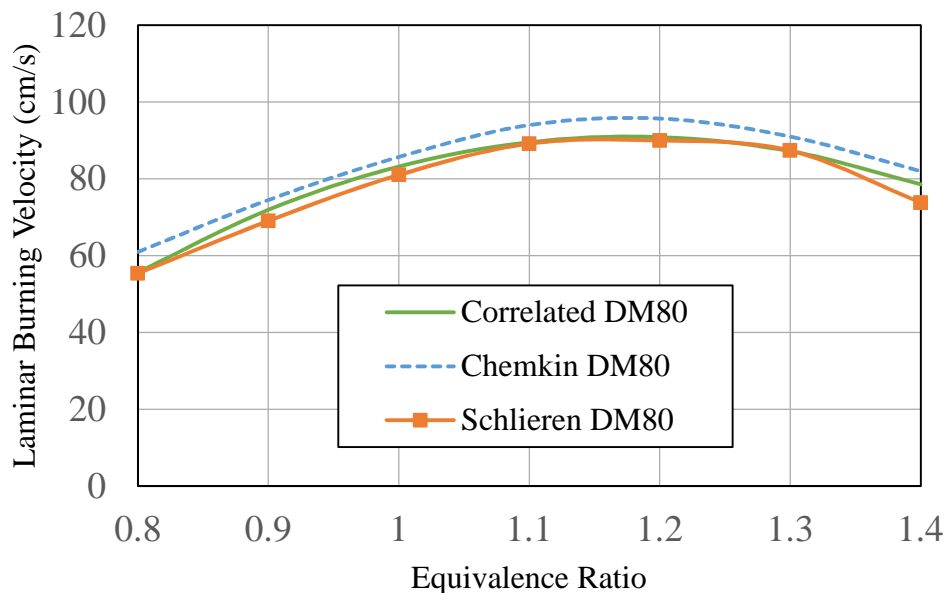


Fig. 10 Burning velocity of natural gas with 80% dissociated methanol (DM80) from correlation, Chemkin and Schlieren at 343 K and 0.3 MPa

Fig.11 shows the laminar burning velocities of DM0, DM40 and DM80 from correlation at 343 K and 0.3 MPa. The peak burning velocities for DM0, DM40 and DM80 are 25.1 cm/s, 40 cm/s and 91 cm/s respectively. At very lean condition with equivalence ratio of 0.8, DM80 still has much higher burning velocity (55.7 cm/s) than the peak value of DM40. For the

stoichiometric mixture, adding 80% dissociate methanol can increase the burning velocity of natural gas by 232%. It can be concluded that dissociated methanol can promote chemical reactions as the burning velocity increases dramatically when dissociated methanol was added. The reason is that both hydrogen and carbon monoxide are chemically active gases and they both have function of promoting combustion. Larger proportion of dissociated methanol leads to higher laminar burning velocity.

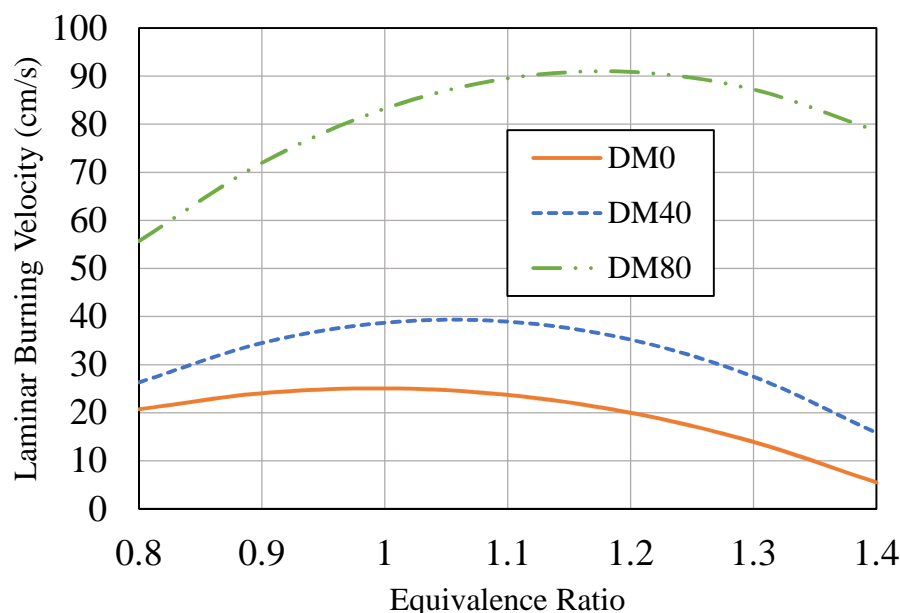


Fig. 11 Burning velocity of natural gas-DM mixtures at 343 K and 0.3 MPa

The peak laminar burning velocity of DM0, DM40 and DM80 occurs at equivalence ratio of 1.0, 1.1 and 1.2 respectively. There is a trend that the peak burning velocity shifts towards the richer side with increasing fraction of dissociated methanol, which shows the similar trend with the results in [27] and [34]. From previous studies [35, 36 and 37], the peak burning velocity of H_2 -air mixtures and syngas ($CO:H_2=99:1$)-air mixtures occurs at around the equivalence ratio of 1.8 and 2 respectively, while the peak laminar burning velocity of CH_4 -air mixtures occurs at around the equivalence ratio of 1. The equivalence ratios of peak burning velocity of CO and H_2 are both higher than that of CH_4 . This can explain why the peak burning velocity shifts towards the richer side when dissociated methanol was added. It can also be seen that the combustible limits are widened with an increasing proportion of dissociated methanol since laminar burning velocity is elevated within the whole equivalence ratio range.

4.4. Effect of Diluent Gas

DM40 was chosen to explore the effect of diluent gas on laminar burning velocity of natural gas-DM mixtures. Fig.12 shows the laminar burning velocity of natural gas-DM mixture (DM40) when nitrogen was added at 343 K and 0.3 MPa.

There is a decrease in laminar burning velocity of DM40-air mixtures when diluted by nitrogen. The peak burning velocity of DM40 is around 40 cm/s but this value decreases to 23.2 cm/s when 5% nitrogen was added (DM40N5). However, the differences of burning velocity among DM40N5, DM40N10 and DM40N15 are much smaller (around 3 cm/s). Adding nitrogen can shift the peak burning velocity towards stoichiometric ratio. The suppression effect of diluent

gas on laminar burning velocity results from the increase in specific heat and hindering the collision of active radicals. For the ultra-lean and ultra-rich mixtures, the amount of active radicals is much lower than the stoichiometric ratio due to the lack of fuel or oxidizer, which can lead to an extremely low collision probability of active radicals. Therefore, the suppression effect of dilution on burning velocity is not very significant at ultra-lean and ultra-rich sides. Therefore adding diluent gas reduces the laminar burning velocity near stoichiometric ratio most significantly compared to the ultra-lean or ultra-rich regions, which is similar to the trend in [12].

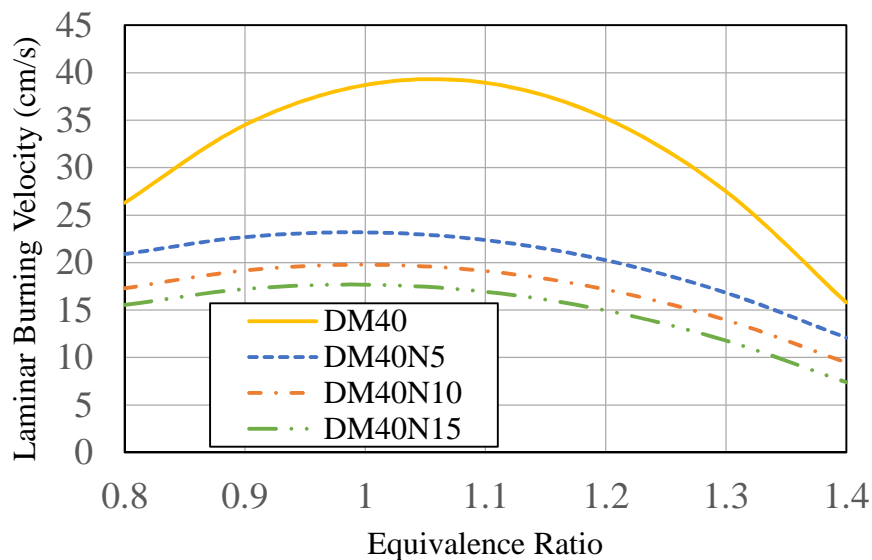


Fig. 12 Burning velocity of natural gas with 40% dissociated methanol (DM40) diluted by nitrogen with different volume fractions at 343 K and 0.3 MPa

4.5. Cellularity

As the cellularity can lead to an over prediction of laminar burning velocity, it is of great significance to detect the onset of cellularity by observing the Schlieren photographs prior to computing the burning velocity from the in-cylinder pressure data [38]. The onset of cellularity of natural gas-DM mixtures with different fractions of dissociated methanol at 343 K and 0.3 MPa is shown in Fig.13. The Schlieren photographs for this condition are shown in Fig.14. There is a trend that higher proportions of dissociated methanol lead to earlier onset of cellularity on flame front. Cellularity occurs when the flame front becomes instable, and this results from local inhomogeneity in the mixture composition within the flame front. The burning rate after the onset of cellularity is no longer uniform, which will result in the uneven flame front. More dissociate methanol can lead to earlier uneven flame front.

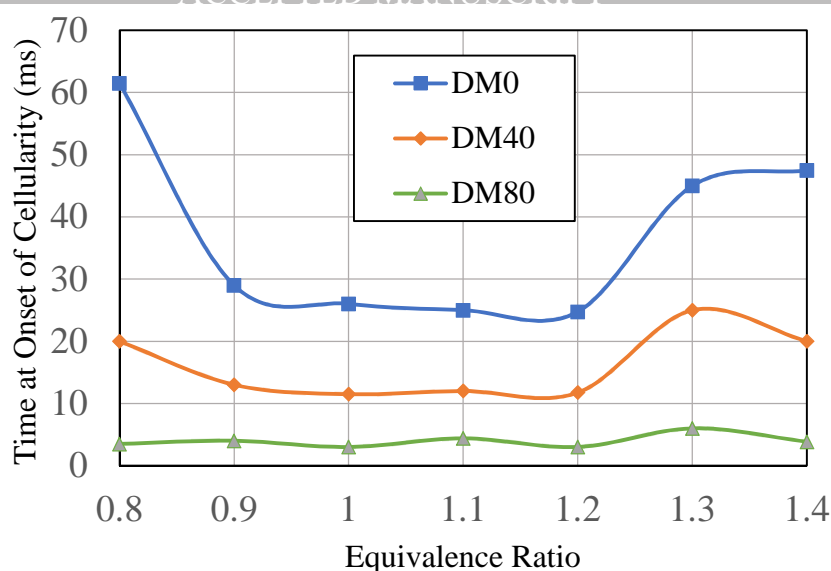


Fig. 13 Onset of cellularity for natural gas with different volume fractions of dissociated methanol at 343 K and 0.3 MPa

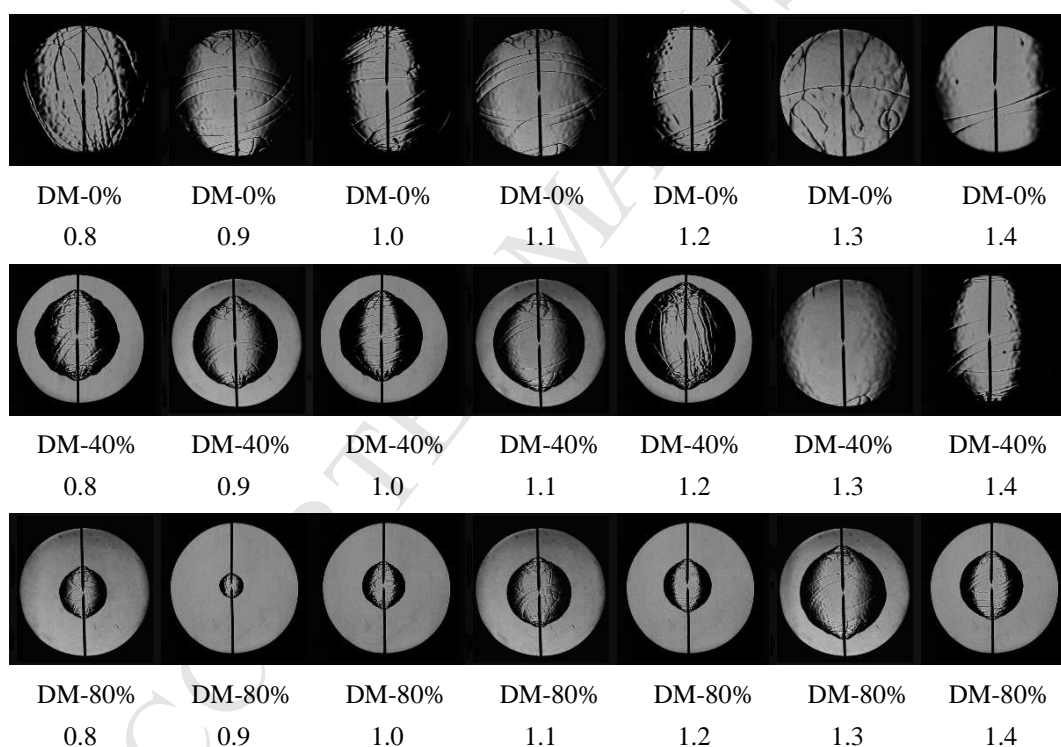


Fig. 14 Schlieren photographs of onset of cellularity at 343 K and 0.3 MPa

4. Conclusions

The laminar burning velocity of natural gas–DM mixtures diluted by nitrogen was calculated via Schlieren photography and correlation of pressure data at initial temperature of 343 K and initial pressure of 0.3 MPa. A numerical model using San Diego mechanism in Chemkin-Pro has been built to predict the laminar burning velocity over wide range of initial temperatures and pressures. The results from these three methods show high agreement with each other. Moreover, cellularity was detected so that the burning velocity data after the onset of cellularity can be excluded to avoid over prediction when using correlation. Key findings are

listed as below:

(1) Higher initial pressures lead to lower laminar burning velocity while higher initial temperatures lead to higher laminar burning velocity. For pure natural gas (DM0), the peak laminar burning velocity occurs close to stoichiometric ratio. This agrees with literature data.

(2) Increasing the fraction of dissociated methanol can increase the laminar burning velocity of natural gas-DM mixtures dramatically. At 0.3 MPa and 343 K, the peak laminar burning velocities of DM0, DM40 and DM80 are 25.1 cm/s, 40 cm/s and 91 cm/s respectively. The equivalence ratio where peak laminar burning velocity occurs shifts towards the richer side with an increasing fraction of dissociated methanol.

(3) Adding nitrogen can lead to dramatic decrease in laminar burning velocity of natural gas-DM mixtures. Increasing the fraction of nitrogen results in lower laminar burning velocity. The equivalence ratio where the peak laminar burning velocity occurs shifts towards stoichiometric ratio when diluted.

(4) The cellularity occurs earlier as the fraction of dissociated methanol increases.

Acknowledgement

None.

References

- [1] Energy Information Administration, US Department of Energy. <https://www.eia.gov/consumption/manufacturing/data/2002/index.php/view=methodology/>, 2002.
- [2] Z. Chen, B. Xu, F. Zhang, et al., Quantitative research on thermodynamic process and efficiency of a LNG heavy-duty engine with high compression ratio and hydrogen enrichment. *Appl. Therm. Eng.* 2017;125:1103–1113.
- [3] J. Liu, X. Duan, Z. Yuan, et al., Experimental study on the performance, combustion and emission characteristics of a high compression ratio heavy-duty spark-ignition engine fuelled with liquefied methane gas and hydrogen blend. *Appl. Therm. Eng.* 2017;124:585–594.
- [4] M. Elsemary, A. Attia, K. Elnagar, et al., Experimental investigation on performance of single cylinder spark ignition engine fueled with hydrogen-gasoline mixture. *Appl. Therm. Eng.* 2016;106:850–854.
- [5] E. Hu, Z. Huang, B. Liu, et al., Experimental study on combustion characteristics of a spark-ignition engine fueled with natural gas-hydrogen blends combining with EGR. *Int. J. Hydrogen Energy* 2009;34:1035–1044.
- [6] Y. Zhang, R. Shang, W. Shen, et al., Extinction limit and near-limit kinetics of lean premixed stretched H₂-CO-air flames. *Int. J. Hydrogen Energy* 2016;41(39):17687- 17694.
- [7] X. Li, Characteristics of methanol decomposed by heat and its application to a SI engine electronically controlled. Tianjin University, 2012.
- [8] B. Peppley, J. Amphlett, L. Kearns, et al., Methanol-steam reforming on Cu/ZnO/Al₂O₃. Part 1: the reaction network. *Appl. Catal. A.* 1999;179:21–29.
- [9] E. Tang, C. Peng, A macro-and microeconomic analysis of coal production in China. *Resources Policy* 2017;51:234–242.
- [10] J. Duan J, F. Liu, B. Sun, Backfire control and power enhancement of a hydrogen internal combustion engine. *Int. J. Hydrogen Energy* 2014;39(9):4581-4589.
- [11] X. Hu, Q. Yu, J. Liu, Chemical effect of CO₂ on the laminar flame speeds of oxy-methane mixtures in the condition of various equivalence ratios and oxygen concentrations. *Int. J. Hydrogen Energy* 2016;41(33):15068-15077.

- [12] X. Zhang, Z. Huang, Z. Zhang, et al., Measurements of laminar burning velocities and flame stability analysis for dissociated methanol–air–diluent mixtures at elevated temperatures and pressures. *Int. J. Hydrogen Energy* 2009;34(11):4862-4875.
- [13] H. Li, G. Li, Z. Sun, et al., Effect of dilution on laminar burning characteristics of $H_2/CO/CO_2$ /air premixed flames with various hydrogen fractions. *Experimental Thermal and Fluid Science* 2016;74:160-168.
- [14] F. Hagos, A. Aziz, S. Sulaiman, et al., Methane enrichment of syngas (H_2/CO) in a spark-ignition direct-injection engine: Combustion, performance and emissions comparison with syngas and Compressed Natural Gas. *Energy* 2015;90:2006–2015.
- [15] F. Hagos, A. Aziz, S. Sulaiman, et al., Effect of fuel injection timing of hydrogen rich syngas augmented with methane in direct-injection spark-ignition engine. *Int. J. Hydrogen Energy* 2017;42:23846–23855.
- [16] F. Lindström, Elmqvist, G. Kalghatgi, et al., An empirical SI combustion model using laminar burning velocity correlations. *SAE* 2005;144(4):833-846.
- [17] J. Farrell, W. Weissman, R. Johnston, et al., Fuel effects on SIDI efficiency and emissions. *SAE* 2003-01-3186, 2003.
- [18] C. Heghes, V. Karbach, J. Warnatz, Evaluation of new data for hydrocarbon kinetics. in: *Proc. Eur. Combust. Meet.*, vol. 2, Louvain-la-Neuve, Belgium, 2005.
- [19] Z. Huang, Y. Zhang, K. Zeng, et al., Measurements of laminar burning velocities for natural gas–hydrogen–air mixtures. *Combust. Flame* 2006;146:302-311.
- [20] X. Zhang, Z. Huang, Z. Zhang, et al., Measurements of laminar burning velocities and flame stability analysis for dissociated methanol–air–diluent mixtures at elevated temperatures and pressures. *Int. J. Hydrogen Energy* 2009;34:4862-4875.
- [21] Z. Chen, On the extraction of laminar flame speed and Markstein length from outwardly propagating spherical flames. *Combust. Flame* 2011;158:291–300.
- [22] G. Markstein, Experimental and theoretical studies of flame-front stability. *Dynam. Curved Fronts* 1988;18:413–423.
- [23] A. Kelley, C.K. Law, Nonlinear effects in the extraction of laminar flame speeds from expanding spherical flames. *Combust. Flame* 2009;156:1844–1851.
- [24] X. Bao, Y. Jiang, H. Xu, et al., Laminar flame characteristics of cyclopentanone at elevated temperatures. *Appl. Energy* 2017;195:671–680.
- [25] O. Manna, M. Mansour, W. Roberts, et al., Laminar burning velocities at elevated pressures for gasoline and gasoline surrogates associated with RON. *Combust. Flame* 2015;162(6): 2311-2321.
- [26] X. Cai, J. Wang, W. Zhang, et al., Effects of oxygen enrichment on laminar burning velocities and Markstein lengths of $CH_4/O_2/N_2$ flames at elevated pressures. *Fuel* 2016;184:466-473.
- [27] Z. Zuo, Y. Pei, J. Qin, et al., Laminar burning characteristics of premixed methane-dissociated methanolair mixtures under lean burn conditions. *Appl. Therm. Eng.* 2018;140: 304-312.
- [28] K. Saeed, R. Stone, The modelling of premixed laminar combustion in a closed vessel. *Combustion Theory and Modelling* 2004;8(4):721-743.
- [29] B. Lewis, V. Guenther, *Combustion, flames and explosions of gases*. London, UK: Academic Press Inc. 1961;8:677-689.
- [30] P. Saxena, F. Williams, Testing a small detailed chemical kinetic mechanism for the

- combustion of hydrogen and carbon monoxide. *Combust. Flame* 2006(145):316-23.
- [31] A. Millán-Merino, E. Fernández-Tarrazo, M. Sánchez-Sanz, et al., A multipurpose reduced mechanism for ethanol combustion. *Combust. Flame*, 2018;193:112-122.
- [32] S. Voss, S. Hartl, C. Hasse, Determination of laminar burning velocities for lean low calorific H_2/N_2 and $H_2/CO/N_2$ gas mixtures. *Int. J. Hydrogen Energy* 2014;39:19810-19817.
- [33] N. Hinton, R. Stone, Laminar burning velocity measurements of methane and carbon dioxide mixtures (biogas) over wide ranging temperatures and pressures. *Fuel* 2014;116: 743-750.
- [34] F. Ren, H. Chu, L. Xiang, et al., Effect of hydrogen addition on the laminar premixed combustion characteristics the main components of natural gas, *Journal of the Energy Institute* (2018), <https://doi.org/10.1016/j.joei.2018.05.011>
- [35] Z. Sun, G. Li, Propagation characteristics of laminar spherical flames within homogeneous hydrogen-air mixtures, *Energy* 116 (2016): 116–127.
- [36] H. Sun, S.I. Yang, G. Jomaas, C.K. Law, High-pressure laminar flame speeds and kinetic modeling of carbon monoxide/hydrogen combustion, *Proc. Combust.Inst.* 31 (2007): 439–446.
- [37] X.J. Gu, M.Z. Haq, M. Lawes, R. Woolley, Laminar burning velocity and markstein lengths of methane-air mixtures, *Combust. Flame* 121 (2000): 41–58.
- [38] K. Liang, R. Stone, Laminar burning velocity measurement of hydrous methanol at elevated temperatures and pressures. *Fuel* 2017;204:206-213.

- The influence of dissociated methanol on laminar flame of natural gas was studied
- Laminar burning velocities were calculated through Schlieren, correlation and Chemkin Pro
- Burning velocity increases with fraction of methanol but decreases with fraction of nitrogen
- Adding more dissociated methanol shifts the peak burning velocity towards richer mixtures
- Lewis number and cellularity were both analysed to show instability of combustion